Second-harmonic generation in planar waveguides of doped silica

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Second-harmonic generation was produced in germanium-doped silica planar waveguides prepared by simultaneous illumination with 1064- and 532-nm laser light. During preparation using prism coupling to specific waveguiding modes, the film-generated second-harmonic intensity grew as a function of preparation time until it saturated. The growth rate and saturation level for p-polarized second-harmonic intensity was an order of magnitude greater than that observed for the s polarization. The efficiency for a 2-cm waveguide length was at least 0.5%. The comparison of experimental results indicates a mechanism for this planar geometry that is similar to that producing harmonic effects in optical fibers.

The efficient production of frequency-doubled light in optical fibers has been observed through optical processing with the fundamental frequency ω (Ref. 1) or additionally seeded with frequency-doubled light 2ω.[2] The process is attributed to the formation of a spatially varying dc field within the fiber, resulting in quasi-phase-matching between ω and the fiber-generated 2ω.[3] Whereas the process for localized modification of optical properties is not fully understood, it has been linked with the atomic and molecular structure associated with dopants and defects within the fiber.[4–6] These effects have been characterized by nonlinear time-dependent growth[1,7] followed by saturation at efficiencies greater than 5%.[8] Because of their one-dimensional nature, no input polarization dependence has been reported for second-harmonic generation (SHG) from fibers[5] or ridge waveguides.[9]

This Letter describes the observation of second-harmonic light generated in two-dimensional waveguiding films that have a composition similar to the optical fibers. We present measurements of second-harmonic growth, saturation, and erasure in specific waveguiding modes of optically processed films. In addition, the dependence of these phenomena on the polarization of the input coupled light is detailed.

The waveguides were produced by ion-beam sputtering of high-purity silica and germanium onto commercial optical-grade fused-silica substrates. The sputter deposition was carried out by an argon-ion beam at a rate of 0.11 nm/s in a partial pressure of 1×10^{-4} Torr of oxygen. The two films used in this study were approximately 2 and 4 μm thick and had an average surface roughness of 1.5 nm. Auger analysis indicated that the germanium content was approximately 6 at. % and was substantially oxidized.

The experimental geometry is shown in Fig. 1. The film was illuminated with 1064-nm (ω) and/or

532-nm (2ω) radiation which was Q switched at 1.22 kHz and mode locked with an average power of as much as 500 mW. This maximum average power produced laser pulses with a peak power of 150 kW. A half-wave plate controlled the polarization of both the ω and 2ω light with a polarization ratio of greater than 98% for both frequencies and polarizations. The laser light was focused with a 23-cm focal-length lens and prism coupled into the film. Scattering from waveguided light at the film–air interface was collected by a fiber-optic waveguide and detected by a Hamamatsu 1P28 photomultiplier tube (PMT). The waveguided light emitted from the film edge was focused by a 10× microscope objective and filtered for detection of only 532-nm light by the PMT. Typically 300–1000 laser pulses were detected by the PMT, averaged by a boxcar, and collected by a computer. Larger SHG signals were detected by a photodiode that was sensitive to ω and 2ω. During measurement of film-generated second-
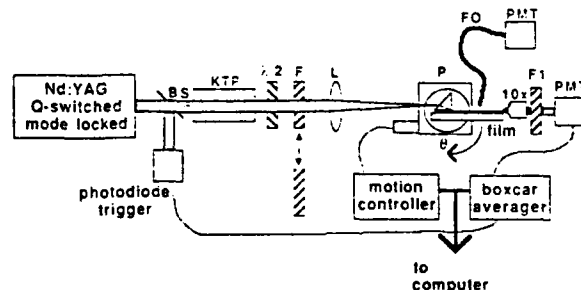

Fig. 1. Experimental setup for thin-film waveguiding experiments. Laser light is coupled into different waveguiding modes by rotation of the film and prism assembly P. To optically prepare the film, filter F is removed to allow illumination of film by ω and 2ω light. To measure film-generated second-harmonic light, F blocks 2ω light. Filter F1 passes only second-harmonic light to the PMT. BS, beam splitter; L, lens; FO, fiber-optic waveguide.
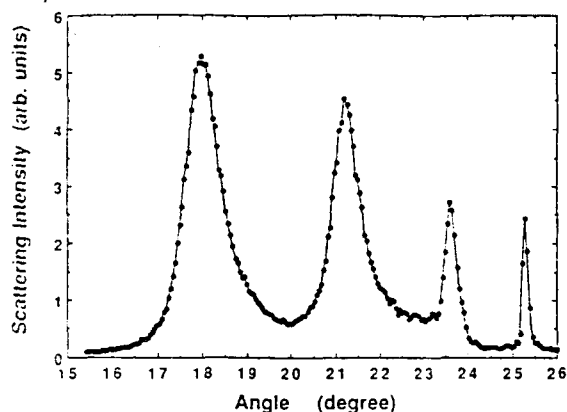
Fig. 2.   Scattering intensity from thin-film waveguiding modes versus the laser light external angle to the coupling prism.   The input beam was s polarized with a 532-nm wavelength.

harmonic intensities, an IR-transmitting filter was placed before the sample assembly to block the 532-nm seed light.

Waveguiding was initiated by coupling ω and/or 2ω light into the film at the right-angle corner of the prism–film interface.   The angles for maximum coupling efficiency were determined by detecting the intensity of scattered light from waveguiding modes as a function of the external coupling angle (Fig. 2). The angular coupling ranges for the lowest-order ω and 2ω modes were 0.64° and 0.35°, respectively. The index of refraction for p-polarized ω and 2ω illumination was calculated[10] to be 1.5234 and 1.5401, respectively, by using the coupling angle for peak scattering intensities.   Small anistropies (0.002) were calculated for s-polarized indices that were slightly greater than the experimental error (0.001). These index values agree with the dispersion measurements obtained by using visible and near-IR transmission measurements.[11]   The waveguide scattering intensity as a function of distance along the waveguiding path indicated that losses in the waveguide were approximately 5 dB/cm.

Initial experiments attempted to produce harmonic generation from films prepared with only 1064-nm illumination.   After preparation times exceeding 8 h, no second-harmonic signal was observed.   In our film experiments, incident peak laser powers of as much as 100 kW were incident upon the coupling prism, but because of the absence of an air gap in this geometry the maximum light coupled into the waveguide was of the order of 1 kW.   The majority of the incident beam was reflected at the prism–film interface.   It would appear that the required threshold of approximately 5-kW peak power to initiate efficient second-harmonic intensities with only the fundamental wavelength[8] was never reached.

When 1064- and 532-nm light was simultaneously coupled into waveguiding modes of a germania-doped silica film, film-generated second-harmonic (FGSH) light grew as a function of time after initiating illumination, as shown in Fig. 3.   The second-harmonic signal was detected by briefly blocking the 532-nm seed beam during the growth process and measuring the

FGSH signal with only IR illumination.   The data indicate an initial quadratic dependence of the second-harmonic intensity for approximately 20 min before becoming nearly linear for as much as 4 h when the second-harmonic intensity saturates.   Multiple paths within the film were prepared, and, in general, saturation occurred between 2 and 10 h.   The time to reach saturation and the maximum FGSH intensity level is highly dependent on the intensity $I(\omega)$ and the ratio of intensities $I(2\omega)/I(\omega)$ coupled into the film. The long preparation times before saturation in our thin films relative to fiber preparation times were attributed to the low preparation power levels within the film, which are due to low coupling efficiency of the laser light.

After saturation, the FGSH effect was stable at room temperature.   Prepared films retained approximately the same FGSH efficiency over a period of 10 days.   One growth study was stopped and restarted 15 h later, with the same efficiency and growth rate as before the interruption.

To ensure that the second harmonic was generated in the film, an output prism coupler was added to intercept both the ω and 2ω light in an unprepared waveguiding path.   After the output-coupled 2ω light reached saturation, the prism was removed, and the second-harmonic light was then observed from the edge of the film, as shown in Fig. 1.   The second-harmonic light again grew as a function of time and saturated.   Because the only change was the addition of a longer, unprepared waveguiding path, we conclude that the additional second-harmonic light was generated within the newly illuminated waveguide path.

At the saturation point, the efficiency of SHG was determined by comparing the collected output power of FGSH with the fundamental light intensity.   After compensating for detector sensitivity, the output at the fundamental frequency was 190 μW and the output at the second harmonic was approximately 1 μW, which gave an efficiency of approximately 0.5%.   The
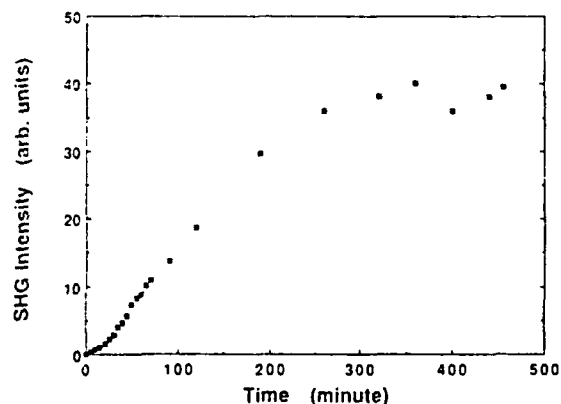

Fig. 3.   Intensity of second-harmonic light generated in the film versus preparation time.   The peak power coupled into the film for preparation was approximately 1 kW for the 1064-nm light and 5 W for the 532-nm light. The FGSH intensity was measured after coupling only 1064-nm radiation into the film with a peak power of 0.8 kW.
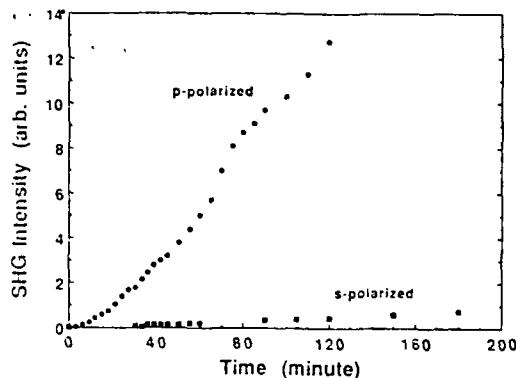
Fig. 4. Intensity of FGSH versus preparation time for s- and p-polarized input preparation beams. The output-coupled intensities of the 1064- and 532-nm preparation beams were the same for the two polarizations.

FGSH light could be observed visually near the saturation intensities.

Both the growth rate and the saturated intensity levels of the FGSH light produced in these doped thin films displayed a dependence on the polarization of the input beams, as illustrated in Fig. 4. The coupling angle for these data was chosen so that the ratio $I(2\omega)/I(\omega)$ was the same for both s and p polarizations, where $I(\omega) \approx 200$ μW and $I(2\omega) \approx 4$ μW. The film was initially illuminated with s-polarized ω and 2ω light, and measurements were made as described above for time-dependent growth measurements. After the film was irradiated with the s-polarized beams for 3 h, a half-wave plate was adjusted to produce p-polarized ω and 2ω illumination. The change in the polarization to p-polarized input light produced a p-polarized FGSH signal that grew an order of magnitude faster than with s-polarized preparation light and a corresponding s-polarized output. The saturated intensities (not shown) of the FGSH light were also an order of magnitude greater for the p-polarization compared with the s-polarized light, but the time for the FGSH intensity to reach saturation was similar for both polarizations. This anisotropy in the polarization dependence may arise when preferentially ejected electrons predicted by some models[12] are effectively confined normal to the plane of the waveguide, while those ejected in the plane would diffuse, further producing a smaller effective χ^2. This model would require either long-lived or multiple photoexcitations to move electrons to the waveguide boundary.

For the experiments described above, FGSH signals were observed only when the input beams propagated along their respective lowest-order wave-guiding modes. The width of the waveguiding modes for ω and 2ω allowed a substantial fraction of the power at the optimal coupling angles to be propagated simultaneously in the film. Typically, the FGSH light was observed after only a few minutes of initiating propagation. In contrast, after 30 min of illumination, no FGSH light was observed for coupling into the second-order mode of the 1064-nm light. The lack of observed FGSH light for this mode

could be due to a mismatch of the optimal coupling angles for the ω and 2ω light, thereby reducing the intensity of copropagating beams. Alternatively, the antisymmetric nature of the second-order 1064-nm mode and the third-order 532-nm mode could produce poor overlap and thus no observable harmonic intensity.

In fiber experiments, grating erasure was demonstrated by a number of techniques, including propagating only the 2ω seed beam through the fiber.[7] We attempted to erase the light-induced grating in the film by initially preparing the doped film with the p-polarized input beam in the lowest-order IR waveguiding mode and subsequently illuminating the film with only 2ω light after removing the 1064-nm input beam. The green seed beam illuminated the film at various 2ω coupling angles. After illumination at 532 nm for periods as long as 6 h, the initial preparation angles and $I(\omega)$ conditions were restored, and no decrease in the FGSH was observed.

In conclusion, we have made what is to our knowledge the first observation of harmonic generation in two-dimensional amorphous doped glass films. These results in thin films are in agreement with observations of growth and saturation of harmonic generation in fibers. The anisotropic response to the input polarization dependence produced by the thin-film geometry offers new insight into the mechanism of the second-harmonic effect. For future studies these films offer the capability of more controlled testing of the optical processing because of the ability to excite and detect specific waveguiding modes through prism coupling. In addition, the simplified geometry allows *in situ* characterization that can identify the microscopic cause of these effects.

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